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Long-Term Goals and Summary of Accomplishments: The goals of this project were to determine links between sediment chemistry, physical properties, and microfabric during the early diagenesis of surface and near-surface, organic-rich sediments. To this end, we provided high resolution characterizations of sediment microfabric, physical properties, chemistry, mineralogy, and organic matter content as well as theoretical work on smectite interlayer hydration. The data collected were used to permit observation of organic matter as it occurs in natural sediments, determine the mechanism for the preservation of organic matter in continental margins and the important role clay minerals play in this regard, to evaluate the impact of the oxygen minimum zone on organic matter abundance and regeneration, and to demonstrate the significant impact organic matter has on sediment physical properties. Mathematical relations were also developed to permit the partitioning of H_2O between hydrous mineral phases in the sediment and intergranular porosity and sediment void ratio and preliminary studies were carried out to examine the potential of the stable isotopes of chlorine for fingerprinting sediment pore waters. In addition, statistical data were accumulated that indicate the range of variability of geotechincal properties on millimeter, centimeter, and kilometer scales for undissected portions of the western North American continental slope.

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Influence of Chemical, Physical, Biological, and Geochemical Processes of Early Diagenesis and Material Exchange Across the Sediment/Seawater Interface in Margin Sediments

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Long-Term Goals and Summary of Accomplishments: The goals of this project were to determine links between sediment chemistry, physical properties, and microfabric during the early diagenesis of surface and near-surface, organic-rich sediments. To this end, we provided high resolution characterizations of sediment microfabric, physical properties, chemistry, mineralogy, and organic matter content as well as theoretical work on smectite interlayer hydration. The data collected were used to permit observation of organic matter as it occurs in natural sediments, determine the mechanism for the preservation of organic matter in continental margins and the important role clay minerals play in this regard, to evaluate the impact of the oxygen minimum zone on organic matter abundance and regeneration, and to demonstrate the significant impact organic matter has on sediment physical properties. Mathematical relations were also developed to permit the partitioning of H_2O between hydrous mineral phases in the sediment and intergranular porosity and sediment void ratio. Preliminary studies were carried out to examine the potential of the stable isotopes of chlorine for fingerprinting sediment pore waters. In addition, statistical data were accumulated that indicate the range of variability of geotechincal properties on millimeter, centimeter, and kilometer scales for undissected portions of the western North American continental slope.

Background: Continental margin sediments are storage sites for highly reactive organic matter and siliciclastic components that intensely interact via chemical, physical, and biological processes. These processes and fluxes of various organic and inorganic sediment components have a profound effect on continental shelf and slope sediment dynamics, consolidation, microfabric, and diagenesis. Organic matter degradation and the dissolution of unstable bioclastic sedimentary components drive chemical changes in surface and near-surface sediments during their first few centimeters of burial, resulting in the development of new sediment chemical and physical properties. Links between the biogeochemical changes in surface and near-surface continental margin sediments and the early evolution of their physical properties are poorly known. Our program sought to identify such links and quantitatively evaluate their impact.

Approach: Sediment samples were collected on two cruises funded by ONR. Sediments were analyzed on vertical and lateral scales of millimeters to centimeters from box and gravity cores along three geologically and oceanographically distinct California continental

margin transects. These analyses provided a unique, high-resolution data set for studies of biogeochemistry and physical properties in continental slope sediments. Sediment samples were augmented by marine snow samples collected from moored and floating sediment trap arrays. These arrays were used to measure fluxes of organic carbon and minerals and to provide material for microfabric analysis. Sediments and sediment trap samples were analyzed for fundamental characteristics such as mass physical properties, grain size, mineralogy, and surface area. In addition, inorganic chemical (i.e., pore water chemistry) and biochemical (organic carbon, carbonate, phosphorous, opal) data sets were determined for sediment samples. Using pore water data and porosity measurements, fluxes of biophilic and other compounds across the sediment-water interface were calculated. In collaboration with Dr. Richard Bennett and the Naval Research Laboratory at the Stennis Space Center, crucial microfabric features (e.g., aggregate structure, organic matter distribution, and dewatering structures) and sediment physical properties (e.g., porosity, water content, and wet and dry bulk density) were also determined. Supporting data on surface and nepheloid layer marine snow fabrics were also collected using wet SEM, TEM, and environmental cell TEM techniques. On the San Luis Obispo transect, in collaboration with Dr. Lisa Levin at Scripps, the impact of benthic organisms and *in situ* fecal pellet generation on organic matter preservation and physical properties was investigated. Both physical property and chemical data were integrated to identify relations between chemical processes and geotechnical properties.

Data and Results: Radiocarbon dates, together with companion $\delta^{13}C$ values from bulk sedimentary carbon and carbonate permitted sediment accumulation rates to be determined from which chemical and physical property changes were interpreted as a function of time and organic matter source. Results show that at the resolution analyzed sedimentation at all three locations has been constant over the past ~15,000 years, except in shallow water sites (< 400 m water depth) where there has been strong current activity, winnowing, and slumping. Sedimentation also appears to be twice as fast (~10 to 70 cm kyr^{-1}) on the two Mendocino transects, which center at longitudes and latitudes of ~ 40°N 50' and 124°W 30' and ~ 40°N 05' and 124°W 24', than on the San Luis Obispo transect (~4 to 40 cm kyr^{-1}), located at ~ 35°N 30' and 121°W 20'. Physical property and pore water chemical data are time independent and were found to be a strong function of sedimentary organic matter content.

Stable carbon isotope data were used to evaluate the relative contributions of continental versus marine organic matter sources. Organic matter in the two northern transects has a significant continental signature (30-50%), especially from sediments in shallow water (up to 70%). In San Luis Obispo sediments, continental contributions to the organic matter is generally less than 15%. Neither area shows preferred preservation of organic matter to be directly correlatable to organic matter source.

Our study of variations in mass physical properties for lateral distances from millimeters to tens of centimeters provide boundary values for further studies of the open continental slope along the western North American continental margin. Our values indicate how much physical property measurements from a site must differ from those at other sites in order to identify statistically significant differences between them. These data also show the extent to which a single set of physical property measurements, taken from

a single core on a relatively undisturbed, typical, uneroded continental slope, can be used to characterize porosity and other geotechnical properties. For typical continental slope sediments, especially those that are fine-grained and close to the sediment-sea water interface, water content and porosity determinations can be highly variable, differing by up to ~58 and 6 %, respectively. Values for maximum and minimum differences in mass physical properties and for values of dispersion for typical coarse- and fine-grained continental slope sediments of different clay contents were determined. Results of this study demonstrate that the short-range variation of geotechnical properties in coarse-grained sediments, such as silty sands, is small compared to that in finer-grained clay-rich sediments. In addition, X-radiographs of undisturbed coarse- and fine-grained sediments, indicate that in relatively undissected areas of the open California continental margin, sedimentation and benthic processes are generally uniform on the kilometer-scale along strike, the major difference being the thickness of sediment accumulated. Satisfactory interpretation of porosity profiles and the origin and significance of even relatively large porosity changes requires, at the very least, organic matter and clay mineralogy determinations and sediment X-radiography.

Sediments with the highest porosity at the sediment-seawater interface have the highest organic matter contents on all transects studied. Surface sediments in the San Luis Obispo area have higher porosity than those in the Mendocino area, reflecting their higher organic matter contents. Results of the pore water analyses indicate that the bacterial degradation of organic matter and the dissolution of bioclastic sediment components provide the driving force for changes in pore water chemistry, with several of the pore water profiles showing oscillatory behavior within the upper 20 cm of the sediment, with pore water maxima at ~5 and 20 cm. These 20 cm of sediment are coincident with the approximate maximum depth of bioturbation by benthic macro- and micro-organisms.

The organic carbon content of sediments deposited on our transects, from which discrete organic particles have been removed, appears to be controlled primarily by its association with clay minerals and other high surface area-to-volume, inorganic sediment constituents. In this regard, different clay suites appear to have different capacities for retaining organic carbon. Relations are well-behaved within a single clay mineral suite, with smectite being associated with the highest organic matter concentrations. This finding has important implications for studies of both sediment physical properties and theoretical and numerical models of sediment particle interaction because coatings of organic matter on clay minerals will significantly affect their surface properties and layer charge potentials, which in turn will impact the fabric and porosity evolution of the sediment.

The organic matter deposited in sediments on the California continental slope appears to occur principally as (1) discrete blebs of organic goo, (2) localized, irregular smears of undifferentiated protoplasmic or extracellular material incorporated into clay flocs, and (3) bacterial cells and their related mucco-polysaccharide networks; not as thin, uniform coatings adsorbed on grain surfaces or infillings of interstices in particles with high degrees of surface roughness. This contradicts the recent "monolayer equivalent" hypothesis of organic carbon preservation to explain the empirical relationship observed between specific sediment surface area and the organic carbon content of continental margin sediments. It is evident from our investigation, that most of the organic matter and microbes in continental margin sediments are localized in occurrence, but intimately associated with layer silicate minerals in the clay

size fraction, with decomposing organic matter adhering to, and microbes attaching themselves to individual clay particles and clay-rich aggregates. This interplay increases sediment cohesion, decreases sediment permeability, and results in a significant slowing of the rate of organic matter decomposition, as organic matter decomposition moves toward the diffusion-controlled limit.

Calculations of the benthic fluxes of biochemical pore water species on the three transects studied demonstrate that continental margin fluids play an important roles in the cycling of biochemical elements between the ocean, atmosphere and crust. This work provides a unique, high resolution data set that links the biogeochemistry of continental margin sediments with its geotechnical and microfabric evolution. These data show a distinctly second order effect of bottom water oxygen content on continental slope organic matter degradation and a first order effect on phosphorous recycling, as well as a strong correlation of organic carbon content and the presence of clay minerals and opal. X-ray diffraction and pore water calculations have also found evidence for the early precipitation of authigenic carbonate fluorapatite, dolomite, and glauconite in the first 20 cm of sediment in all CALMAR transects. These authigenic minerals, if acting as cementing agents, undoubtedly will affect sediment physical properties. TEM scans for their presence are proposed.

Theoretical work on the properties of the mineral smectite, a hydrated clay mineral in which easily exchangeable intracrystalline H_2O occurs, provided the basis for the mathematical treatment of water content, porosity, bulk density, void ratio, and other water content-dependent geotechnical properties. This work provides both the equations and laboratory protocol for partitioning H_2O between smectite interlayers and intergranular pore spaces in the sediment. Application of this correction permits the true porosity and compaction history of the sediment to be determined.

Scientific Impact: This grant has provided funding which has permitted us to make a number of important breakthroughs in understanding the early diagenesis of continental margin sediments and the behavior of two of their most reactive components: clay minerals and organic matter. These results can be applied to studies of the cycling of biogeochemical aqueous species between the sediment and overlying water column and the flux of these components; studies of sediment physical property response and interpretation, and organic matter preservation and distribution. In addition to academic and engineering applications, sedimentary organic matter is an important scavenger of metals and other types of pollutants from seawater, therefore, our results could also be important from an environmental point of view. They may shed light on the location and physical micro-environments of major, active, adsorptive sites for metals and toxic pollutants. The direct observation of the textural interplay between organic matter, microbes, and clay minerals can be used to improve the design of experiments and models implemented for the purpose of identifying and monitoring processes relating to the sediment uptake and release of toxic organic-chlelated compounds in the marine environment.

This research is compatible with Naval engineering, as well as applied research and development programs involved with system interactions in coastal and shallow water marine environments.

Publications Supported by ONR Grant # N00014-92-J-1225

Brown, K.M. and Ransom, B. (1996) Porosity corrections for smectite-rich sediments: Impact on studies of compaction, fluid generation, and tectonic history. *Geology*, v. 24, 843-846.

Lavoie, D. M., Little, B. J., Ray, R. I., Bennett, R. H., Lambert, M. W., Asper, V. and Baerwald, R. J. (1995) Environmental scanning electron microscopy of marine aggregates. *Jour. Microscopy*, v. 178, 101-106.

Lavoie, D. M., Baerwald, R. J., Hulbert, M. H. and Bennett, R. H. (1996) A drinking straw mini-corer for sediments. *J. Sedimentary. Research*, v. 66, p. 1030.

Magenheim, A.J., Spivack, A.J., Volpe, C. and Ransom, B. (1994) Precise low-level determination of stable Cl isotopic ratios in silicates and aerosol samples. *Geochimica et Cosmochimica Acta*, v. 58, 3117-3123.

Ransom, B., Bennett, R. H. and Baerwald, R; Shea, K. (1997) *In Situ* Organic Matter in Recent Marine Sediments: A TEM Investigation of Organic Matter Preservation on Continental Margins and the "Monolayer" Hypothesis. *Marine Geology* v. 38, 1-9.

Ransom, B., Spivack, A.J. and Kastner, M.(1995) Stable chlorine isotopes in subduction zone pore waters: Implications for fluid-rock reactions and the cycling of chlorine. *Geology*, v. 23, 715-718.

Publications in Presently in Press or Review Supported by ONR Grant # N00014-92-J-1225

Fitts, T.G., Brown, K.M., Ransom, B. Correcting porosity in smectite-rich sediments: A method using bulk density data compatible with logging while drilling (LWD) data. Geotechnique (in review).

Herbert, T.D., Schuffert, J., Lange, K., Weinheimer, A. and Herguera, J.C. A core-top calibration of the alkenone undersaturation index along the California margin. *Paleoeceanographpy* (in press).

Hulbert, M.H., Bennett, R.H., Baerwald, R.J., Lavoie, D.M. and Long, R.L. Observations of the sediment-water interface: Marine and fresh water environments. *Clay Minerals* (in review).

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Sawyer, W., Ransom, B. and Bennett, R.H. Geotechnical Property Variability of Continental Margin Sediments: High-Resolution, Vertical and Lateral Data from the Northern California Slope. Marine Georesources and Geotechnology (in press).

Shea, K.F., Ransom, B., Burkett, P.J., Bennett, R.H., Baerwald, R. TEM comparison of pelagic and nepheloid layer marine snow: Implications for carbon cycling on the California margin. Marine Geology (in review).

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PUBLICATIONS

(12) Papers which were supported by this grant that are presently published, in review, or in press as of September 1, 1997:

Brown, K.M. and Ransom, B. (1996) Porosity corrections for smectite-rich sediments: Impact on studies of compaction, fluid generation, and tectonic history. *Geology*, v. 24, 843-846.

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Magenheim, A.J., Spivack, A.J., Volpe, C. and Ransom, B. (1994) Precise low-level determination of stable Cl isotopic ratios in silicates and aerosol samples. *Geochimica et Cosmochimica Acta*, v. 58, 3117-3123.

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Ransom, B., Kim, D., Kastner, M. and Wainwright, S. Organic Matter Preservation on Continental Slopes: Importance of Mineralogy and Surface Area. *Geochemica et Cosmochimica Acta* (in press).

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Shea, K.F., Ransom, B., Burkett, P.J., Bennett, R.H., Baerwald, R. TEM comparison of pelagic and nepheloid layer marine snow: Implications for carbon cycling on the California margin. Marine Geology (in review).

(8) Papers in Preparation:

Bennett, R.H., Ransom, B., Kastner, M., Sawyer, W., Baerwald, R.J., Lambert, M.W. and Burkett, P.J. Early marine sediment diagenesis: Properties and processes, California continental margin (to be submitted to *Marine Geology*).

Kastner, M., Ransom, B., Bennett, R.H., Kim. D. The interplay of chemical and physical processes in early sediment diagenesis (to be submitted to Marine Chemistry).

Kastner, M., Kim, D., and Schuffert, J. Formation of Authigenic Carbonate Fluorapatite (Francolite) in California Continental Slope Sediments (to be submitted to *Geochimica et Cosmochimica Acta*).

Ransom, B. Lambert, M. and Bennett, R.H., Organic matter composition on the California margin and its impact on organic carbon preservation (to be submitted to *Marine Geology*).

Ransom, B. and Kastner, M. Leaching or organic matter during grain size separation: Results of mass balance. (to be submitted to *Deep Sea Research*).

Ransom, B., Kim, D., and Kastner, M., Rates of sediment and organic matter accumulation on organic carbon preservation, benthic biochemical fluxes, and sediment porosity: Example from the California margin (to be submitted to *Geochimica et Cosmochimica Acta*).

Ransom, B. Mechanisms of formation of mixed-layered clays: Implications for thermodynamic properties, experimental studies, and clay mineral classification (to be submitted to *American Mineralogist*)

Fitts, T. and Ransom, B. The thermodynamic properties of interlayer H2O in smectite: Experimental consideration and approaches. (to be submitted to *Geochimica Cosmochimica Actas*)

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Kim, D. (1996) *Biochemical Cycling of Carbon, Phosphorous, and Silica in California Continental Slope Sediments.* Ph.D. dissertation, Scripps Institution of Oceanography, University of California at San Diego, La Jolla, California.

Shea, K. (1997) Masters Thesis, University of Southern Mississippi (in preparation)

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Bennett, R.H., Kastner, M., Ransom, B., Herbert, T., Kim, D., Fitts, T., Robertson, G., Moreno, M., Baerwald, R., Asper, V., Lambert, M., Lavoie, D., Burkett, P.J., and Kennedy, C. (1994) Microfabric, physical properties, and biogeochemistry of

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(1) Book Chapters:

Lavoie, D. M. (1996) Examination of hydrated, non-conducting biofilms in ESEM (in Proceedings in Electron Microscopy) (in press).

(2) Workshop/Reports:

Bennett, R.H. (1991) ONR Marine Sediment Biogeochemistry Workshop: Biogeochemical processes driving sediment microstructure development and geotechnical properties variability. 50 p.

Bennett, R. H., Kastner, M., Baerwald, R. J., Ransom, B., Sawyer, W., Lambert, M. W., Olsen, H., Hulbert, M. H. (1996) Early Marine Sediment Diagenesis: Properties and Processes, California Continental Margin. Report to Office of Naval Research, Stennis Space Center: Contract number N00014-96-C-6034. 98 p.

ORAL PRESENTATIONS

(i.e., 12 abstract presentations + 8 non-abstract presentations)

Bennett, R.H. (1995) Modeling microfabric of fine-grained marine sediments: Recommendations for future research". ONR/URI Sediment Geoacoustical and Geotechnical Constitutive Modeling Workshop, Narraganset, RI

Brown, K.M., Fitts, T., Tyron, M. and Ransom, B. (1996) An experimental study of the physical and chemical property changes occurring during the smectite to illite reaction. American Association of Petroleum Geologists Annual Meeting Official, San Diego, v. 5. p. A19.

Fitts, T. Brown, K.M., Tyron, M., and Ransom, B. (1996) Measurement of porosity and smectite hydration state under deviatoric stress conditions: Implications for water partitioning between pores and minerals in sedimentary basins (American Association of Petroleum Geologists Annual Meeting Official, San Diego, v. 5. p. A46.

Herbert, T.D., Stott, L.D. and Herguera, J.C. (1995) Glacial-interglacial SST reconstruction along the California margin from alkenone undersaturation indices, EOS V. 76, F 296.

Herbert, T. D., Schuffert, J., Thomas, D., Lange, K., Weinheimer, A. and Herguera, J.C. (1996) Alkenones in California margin sediments: Indices of sea surface temperature and phytoplankton productivity. 1996 Ocean Sciences Meeting, EOS, v. 76, p. OS201.

Kastner, M. and Ransom. B. (1995) Chloride concentration and chlorine stable isotope composition of fluids in subduction zones. Geologic Association of Canada/Mineralogical Association of Canada Annual Meeting Abstract volume, Victoria, British Columbia.

Lambert, M.W., Bennett, R. H. and Kastner, M. (1993) Variation in terrestrial input to sea floor muds off Cape Mendocino, California. Annual Clay Mineral Society Meeting, San Diego, CA.

Olsen, H.W. and Bennett, R.H. (1996) Hydrologic properties of marine sediments near the sea-water interface and their geologic controls. 1996 International Offshore and Polar Engineering Conference, Los Angeles.

Ransom, B. (1995) The new clay mineralogy, A revolution whose time has come. Goldschmidt Conference in Geochemistry Program with Abstracts, 1995, College Station, Pennsylvania. p. 82.

Ransom, B. (1995) Illite and illitic-mixed-layered clay formation in shales: Dynamic crystallization model. Geological Society of America Abstracts and Programs, v. 27, p. A461.

Ransom, B., Kastner, M., Kim, D., Bennett, R.H., Lambert, M. (1996) Relations Between Organic Matter, Mineralogy, and Physical Properties of Sediments on the California Continental Margin. EOS, Transactions of the American Geophysical Union, v. 76, no. 3, p. OS200.

Ransom, B. Kastner, M., Kim, D., Bennett, R.H. and Lambert, M. (1996) Relations Between Organic Matter, Mineralogy, and Physical Properties of Sediments on the California Continental Margin (accepted for 1996 AGU Ocean Sciences Meeting in San Diego).

Ransom, B., Kastner, M. and Bennett, R.H. (1996) Organic matter in California continental margin sediments: Where is it, how does it get there, and how much is preserved? Geological Society of America Abstracts and Programs, v. 28, p. T28.

Ransom, B. and Kastner, M. Organic Matter Preservation in Continental Margin Sediments: Surface Area, Mineralogy, and the Monolayer Hypothesis. (1997) Geological Society of America, to be presented at the fall meeting, October, 1997.

Ransom, B. (1993) Smectite dehydration: What really goes on between the sheets. Scripps Institution of Oceanography Geochemistry Seminar

Ransom, B. (1994) Clays, Chlorine, and Convergent Margins, University of California at Los Angeles, Rubey Colloquium.

Ransom B. (1994) Out of Chaos: The New Clay Mineralogy, University of California at Los Angeles, Seminar Series

Ransom, B. (1996) Clays, Chlorine, and Convergent Margins, University of Colorado at Boulder, Departmental Colloquium)

Ransom, B. (1996) Organic Matter in Continental Margins Sediments: How Does It Get There and Why Does It Stay, Arizona State University, Departmental Colloquium

Ransom, B. (1996) Mixed-layered Clays and the New Clay Mineralogy, Arizona State University, Departmental Colloquium.

PATENTS

Hydrodynamic dampening system for the precise measurement of dynamic sediment pore water pressures. (R.H. Bennett, application submitted)

(20) INVOLVED ACADEMIC PERSONNELL: 6 women, 3 minorities

(3) Undergraduate Students: (Caucasian)

Female: Sonya Wainwright (SIO)

Male: Tobin Lee (SIO), ethnic group: Oriental
Anthony Moreno, ethnic group: Hispanic

(3) Graduate students: (except where noted, participants are Caucasian)

Male: Dongseon Kim (SIO), ethnic group: Oriental
Kevin F. Shea (NRL)
Kingsley H. McCrocklin (NRL)

(3) Post Doctoral Fellows: (Caucasian)

Female: Barbara Ransom (SIO)

Male: Michael Lambert (NRL)
Steven Chambers (SIO)

(3) Staff: (Caucasian)

Female: Gretchen Robertson (SIO)

Patti Jo Burkett (NRL)
Male: Conrad Kennedy (NRL)

(8) University and Other Professionals: (Caucasian)
Female: Miriam Kastner, (SIO)
Lisa Levin (SIO)
Male: Timothy Herbert (Brown University)
Arndt Schimmelmann (Indiana University)
Hal Olsen (USGS)
Ray Ferrell (Louisiana State University)
Roy Baerwald (University of New Orleans)
Richard Jahnke (Skidaway Marine Station)
Vernon Asper (University of Southern Mississippi)

P.I. SERVICE ON COMMITTEE AND PANELS OUTSIDE OF SIO

Miriam Kastner:

Member of Editorial Board: Aquatic Geochemistry - 1994-present

Sedimentary and Geochemical Processes Panel Member, Ocean Drilling Program - 1995-present

Member of Honorary Fellows Committee, Geological Society of America - 1996-present

Member of Goldschmidt Medal Committee - The Geochemical Society - 1996-present

Coordinating Board of the University of California, Water Resources Center - 1995-present

Member of Sedimentolgy and Geochemical Processes Panel, Ocean Drilling Project - 1995

Editor: Earth and Planetary Science letters - 1988-1995

Member of Editorial Board: Annual Review of Earth and Planetary Science Letters - 1991-1995

Counselor: Geochemical Society - 1990-1994

P.I. RECENT HONORS AND AWARDS

Miriam Kastner:

Fellow of the American Geophysical Union - 1997
JOI/USSAC Distinguished Lecturer - 1995-1996
Phi Beta Kappa visiting scholar - 1993-1994
Invited Participant: ONR Marine Sediment Biogeochemistry Workshop - 1994
Fellow, American Association for the Advancement of Science - 1993
ONR Science Educators Award - 1991